

Coherent control for the spherical symmetric box potential in short and intensive XUV laser fields

I. F. Barna¹ and P. Dombi²

¹ *Atomic Energy Research Institute of the Hungarian Academy of Sciences,
(KFKI-AEKI), H-1525 Budapest,
P.O. Box 49, Hungary*

² *Research Institute for Solid State Physics and
Optics of the Hungarian Academy of Sciences,
(KFKI-SZFKI) H-1525 Budapest,
P.O. Box 49, Hungary*

(Dated: February 5, 2008)

Abstract

Coherent control calculations are presented for a spherically symmetric box potential for non-resonant two photon transition probabilities. With the help of a genetic algorithm (GA) the population of the excited states are maximized and minimized. The external driving field is a superposition of three intensive extreme ultraviolet (XUV) linearly polarized laser pulses with different frequencies in the femtosecond duration range. We solved the quantum mechanical problem within the dipole approximation. Our investigation clearly shows that the dynamics of the electron current has a strong correlation with the optimized and neutralizing pulse shape.

PACS numbers: 32.80.Qk, 32.80.Fb, 32.80.Wr

I. INTRODUCTION

Coherent control has become a routine procedure in physics and chemistry to optimize and govern light-matter interaction processes in atomic and molecular systems [1, 2]. However, widespread coherent control methods are limited to the shaping of visible or near-IR femtosecond laser pulses and to controlling corresponding transitions induced by them. There is, however, enormous future potential in this method. For example, extreme ultraviolet (XUV) pulses can be used to investigate hyper-fast inner shell processes in atoms and molecules and this possibility naturally raises the question whether a certain control can be also exercised by shaping the interacting attosecond waveforms. One of us has tried to answer this question by theoretically investigating a non-resonant two-photon transition in He (1s1s) - (1s3s) with shaped XUV pulses [3]. Even though the answer was positive these calculations lacked insight into the fundamental physical background of the control process. Therefore, in this paper we investigate a more simple model system and analyze the control process for this parameter regime further.

With the proliferation of tabletop, Ti:sapphire-laser-driven XUV sources based on high harmonic generation, experimental tools have also become available even to university groups to carry out similar coherent control like experiments. This can be achieved in two ways: i) either by phase shaping the XUV beam after the high harmonic generation process to achieve the desired temporal shaping effect [4] or ii) by exercising the well-known coherent control methods on the infrared laser pulse generating the harmonics. Even though, to our knowledge, this latter option has mainly been used to maximize harmonic conversion efficiency [5, 6, 7], either by temporal or by spatial shaping of the genetic IR pulse. Option i) is also attractive since it provides a more direct control method and first pioneering experiments by Strasser *et al.* [4] have shown that this scheme is suitable for controlling coherent transients in a He atom. The demonstrated method also has the advantage of direct applicability to free-electron-laser beam-lines currently being developed. These novel light sources will also induce research along these lines in a broad spectral domain currently unavailable to laser-driven XUV/X-ray sources.

For the above-mentioned motivations we investigate the control process further with numerical tools for a more simple model system, a spherical box potential, and a simplified ansatz for the genetic algorithm. The motion of an electron was investigated in a spherically

symmetric square well potential driven by a linearly polarized extreme XUV laser pulse. We solved the time-dependent Schrödinger equation with our simplified coupled-channel method which was successfully applied for more complex laser-atom interaction problems [8]. Following the conventional experimental setups we apply the genetic algorithm (GA) [9]. as optimization procedure to create the best interacting or most indifferent pulses (we call it neutralization) for state selective excitation. Our results show that the wave packet dynamic, the center-of-mass of the electron current is strongly correlated with the shape of the laser pulse, which gives a physical interpretation for the control mechanism, at least for this model potential problem. Section 2 shortly outline the theoretical background of our used model, followed by a compact description of the GA. Section 3 present our results with an explanation. Atomic units [a.u.] are used through the paper unless otherwise indicated.

II. THEORY

We solve the general time-dependent problem with our simplified coupled-channel approach to describe controlled laser driven excitation processes in the spherically symmetric box potential . The original method can be found in our former studies [8],[10]. For the expansion coefficients of the time-dependent wavefunction the following differential equation system holds

$$\frac{da_k(t)}{dt} = -i \sum_{j=1}^N V_{kj}(t) e^{i(E_k - E_j)t} a_j(t) \quad (k = 1, \dots, N) \quad (1)$$

where E_k and E_i are the eigenvalues of the box potential, and will be specified later. The coupling matrix elements

$$V_{kj}(t) = \langle \Phi_k | \hat{V}(t) | \Phi_j \rangle. \quad (2)$$

are taken with the well known eigenfunctions of the box potential. The probabilities for transitions into final excited states j after the pulse are simply given by

$$P_j = |a_j(t = T)|^2 \quad (3)$$

where T is the duration of the pulse. To get the total excitation probability the corresponding channels P_j must be summed up. When a state selective excitation probability is controlled then the corresponding channel is considered only. We have to mention, that the box potential has a continuous spectra as well, which can interpreted as an ionization spectra.

In the following we concentrate on non-resonant two-photon excitation processes and neglect three-photon ionization yields which have negligible contributions in similar atomic systems [3]. We restrict ourselves to linearly polarized laser pulses parallel to the z-axis. The length gauge with the dipole approximation is applied

$$V(t) = -\mathbf{E}(t) \cdot \mathbf{r}. \quad (4)$$

To understand the control mechanism we took a simple model, and investigated the three-dimensional spherically symmetric square-well potential. With the help of the width 'b' and the depth '-V₀' (which are the only two parameters of this potential) the number of the bound states can be fixed. We tune these parameters in such a way (b= 5 a.u., V₀= 5 a.u.) that only four bound states exist. The four state have different angular momenta from zero up to three. A detailed analysis of the problem can be found in any textbook [11]. The wave functions inside the box potential are the well known spherical Bessel functions, and the energies can be found as solutions of different transcendental equations. The four bound states have the following energies: $E_{\ell=0} = -3.6$ a.u., $E_{\ell=1} = -1.85$ a.u., $E_{\ell=2} = -0.36$ a.u., $E_{\ell=3} = -0.05$ a.u.

For external driving field strength we add three different frequencies and use a sin² envelope,

$$\vec{E}(t) = E_n \cdot \sin^2\left(\frac{\pi t}{T}\right) [a_1 \sin(\omega_1 t + \delta_1) + a_2 \sin(\omega_2 t + \delta_2) + a_3 \sin(\omega_3 t + \delta_3)] \vec{e}_z. \quad (5)$$

where the frequencies are fixed and the three amplitudes $a_{1,2,3}$ and phases $\delta_{1,2,3}$ are the free parameters optimized through the GA [9]. For ω_1 we took a quasi resonant two-photon frequency: $(E_{\ell=2} - E_{\ell=0})/2 \approx \omega_1 = 1.52$ a.u. The resonant frequency is 1.62 a.u. and when ω_1 is closer to the resonance then all the pulses excite the system with a large probability ($P_2 > \text{ten percent range}$), and the electron dynamics between the optimized and neutralized cases have the same properties. On the other hand if ω_1 is much farther from resonance then the optimization can not give us enough excitation, and the system remains practically in ground state. For the two other frequencies we took $\omega_2 = 1.40$ a.u. and $\omega_3 = 1.85$ a.u. The GA freely varies the phases $|\delta_{1,2,3}| \leq \pi$ and the amplitudes $|a_{1,2,3}| \leq 2$ which allows combinations where the contribution of the quasi resonant frequency is suppressed letting the off-resonant frequencies to evolve their effect. Contrary to the widely used phase

modulation, this pulse fabrication mechanism does not conserve pulse energy, that is why all pulses have to be renormalized to a reference pulse

$$\int_0^T |\vec{E}(t)|^2 dt = 25.13 \text{ a.u.} \quad (6)$$

in this pulse we took the quasi resonant frequency only, $\omega = 1.52$ a.u., with a large field strength $E_0 = 0.6$ a.u., and $T = 400.53$ a.u. pulse duration and E_n is the normalization constant eq. (5). This parameter set gives us massive excitation probabilities which is needed for the further investigation.

For time propagation we use a Runge-Kutta-Fehlberg method of fifth order embedding an automatic time step regulation. As an optimization procedure we used the celebrated genetic algorithm which works in the following way. GA represents each possible solution, or individual, with a string of bits, termed a chromosome. For example two possible phases are represented as [01101101]. (For the sake of clarity we assume that each parameter can only take $2^3 = 8$ values in this example.) The first generation of individuals is selected randomly. Typically, we use a population of size of 20 which is about a factor of three larger than the number of the optimisable variables. For each generation, the following steps are carried out: (i) all the individuals are evaluated and assigned a fitness value. In our case it means that we calculate the ionization or state selective excitation probability resulting from each parameter configuration and call it fitness value. The next generation of individuals is chosen by applying three GA operators selection, mutation and crossover. (ii) The selection operator chooses which of the individuals from the present generation will be transferred to the next generation. The individuals are ranked according to their fitness, and then selected randomly with a certain probability based on the fitness. A very fit individual thus receives a high probability and can be selected many times, while low-fitness individuals may not be selected at all. (iii) The mutation operator, which is used very seldomly selects a few individuals and replaces one (randomly selected) bit in a chromosome randomly by 0 or 1 (e.g. creating the chromosomes [11010100] from [11000000].) (iv) The crossover operator takes two individuals at a time and exchanges part of their chromosomes. For example two chromosomes [01101000] and [01001110] can create the chromosomes [01101110] and [01001000]. The use of the mutation and crossover operators ensures that the GA does not become in a local minimum or maximum. The fittest individuals, however always survives to the next generation what is called elitism. Steps (i)-(iv) are then repeated until no new

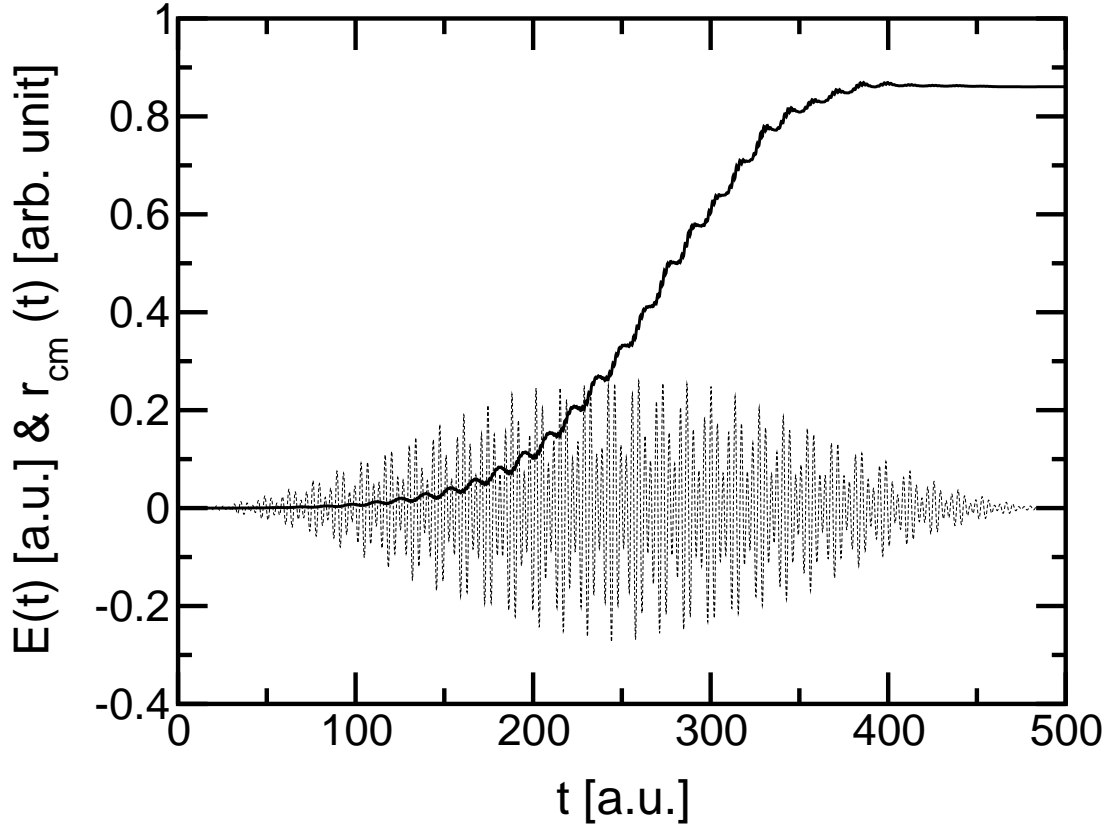


FIG. 1: The electric field strength $E(t)$ of the optimized pulse (dashed line) with the corresponding center of mass of the electron current $r_{cm}(t)$ solid line)

solutions appear between two consecutive generations. In the optimizations presented in this work, the GA typically converges after 40-70 generations.

III. RESULTS

The laser field has six free parameters that have to be optimized through the control process. The GA needs further, technical parameters [9]. We used 20 different pulses per generation (population size) and let the process run through 60 generation to achieve convergence. This means that $20 \times 60 = 1200$ different pulses were checked to find the most favorable. For the permutation probability ($\approx 1/\text{population size}$) = 0.05, for crossover probability = 0.4, and for creeping probability = 0.12 values were used giving us good convergence and gain matching the commendation of the routine [9]. We found the algorithm stable and robust against slight parameter variations.

The GA makes it possible to carry out two different kinds of optimization calculations, first the maximization of the excitation probabilities (we call it optimization) and secondly minimization (we call it neutralization) when the transition probabilities are minimized. Both of these calculations had to be done, to interpret the control mechanism. Neutralization calculation may attract large interest in future FEL experiments where on one side the maximal field strength is used but on the other side the field must not interact with the resonator causing any damage. The optimized P_2 transition probability for the pulse given above is 2.3×10^{-3} , whereas for the neutralized probability it is 2.8×10^{-5} which is a factor of 82 difference. The parameters of the optimized pulse are: $a_{1,2,3}[1.57, 0.68, 1.38]$ $\delta_{1,2,3}[0.45, -0.88, 0.52]$ and for the neutralizing pulse: $a_{1,2,3}[1.17, 1.68, 0.82]$, $\delta_{1,2,3}[-0.52, 0.66, 0.12]$

To find some physical interpretation for our control results, we investigated the dynamics of the electron wave packet. We calculated the time-dependent current of the electron through the following formula:

$$\mathbf{j}(\mathbf{r}, t) = \Psi(\mathbf{r}, t)^* \vec{\nabla} \Psi(\mathbf{r}, t) \quad (7)$$

for the time-dependent wave function we used the usual form:

$$\Psi(\mathbf{r}, t) = \sum_{\ell=0}^3 a_{\ell}(t) \Phi_{\ell}(\mathbf{r}) e^{-iE_{\ell}t} \quad (8)$$

where

$$\Phi_{\ell}(\mathbf{r}) = \left(\frac{\pi}{2r}\right)^{\frac{1}{2}} \cdot J_{\ell+\frac{1}{2}}(rk) Y_{\ell,0}(\theta, \varphi). \quad (9)$$

The radial part $\sqrt{\pi/2r} J_{\ell+0.5}(rk)$ is the spherical Bessel function of the first kind, (usually noted with $j_{\ell}(rk)$ which may leads to miss-understanding in our case using the same letter for electron current) and $J_{\ell+\frac{1}{2}}(rk)$ is the ordinary Bessel functions of half-odd-integer order, and $Y_{\ell,0}(\theta, \varphi)$ is the usual spherical harmonic.

The current is a complicated quick oscillating function, and it is difficult to observe its correlation with the laser field, that is why we introduce the center-of-mass coordinate of the electron current

$$r_{cm}(t) = \frac{\int j(r, t) \cdot r dr}{\int j(r, t) dr} \quad (10)$$

which is a pure time-dependent number. We concentrate on the radial current component only, skipping the vector notation. It is easy to see, that the center-of-mass of the current is a bound function of time. Before the laser pulse - without any external field - the system

is in the $\ell = 0$ ground state with a wave function proportional to $\frac{\sin(rk)}{rk}$. This function has maximum at zero and has a strong decay, hence the center-of-mass coordinate is close to zero. On the other hand the wave functions of the excited states are higher order Bessel functions having a zero at the origin and maximum at $r = 5.0$ a.u. Inside a laser pulse the electron is in a mixed quantum state. With a linear scaling we may identify the minimum of the current to zero and the maximum to a number comparable with field peak intensity. Figure 1 shows the electric field strength of the optimized pulse together with the corresponding electron current function. Beyond the \sin^2 envelope shape and the carrier oscillation an extra modulation can be found with a quasi periodic time $\tau \approx 14$ a.u. This property of the pulse is a beat phenomena. The well-known addition theorem says:

$$\sin(\alpha) + \sin(\beta) = 2 \sin\left(\frac{\alpha + \beta}{2}\right) \cos\left(\frac{\alpha - \beta}{2}\right) \quad (11)$$

which means that adding two different frequencies with the same amplitude and the same phase gives us a new completely amplitude modulated signal with the beat time of: $\tau_{beat} = \frac{\pi}{|\alpha - \beta|/2}$. By checking the parameters of the optimized pulse, we found that ω_2 and ω_3 have approximately the same amplitude and the same phase, giving us $\tau = 13.96$ a.u. The amplitude modulation is not maximal due to the existence of ω_1 .

Let's investigate the current now. It is clear to see that for $t > 150$ a.u. the current continuously gains between two neighboring beat oscillation minima, and has a short plateau at the vicinity of the minima. This behavior can be explained as a resonance phenomenon, where the electron absorbs energy from the laser pulse at each intensity growth. The time-dependent center-off-mass of the electron current shows two kinds of oscillations. The slower oscillation follows the envelope of the pulse, (we call it envelope oscillation) and a much quicker but smaller oscillation follows the carrier of the pulse (we call it carrier oscillation). The work of Kosloff *et al.*[12] prove that there is a $\pi/2$ phase shift between the optimal pulse and the time dependent wave function overlap. Figure 2 shows a magnified part of the field strength of the optimized pulse together with the overlap of the time-dependent ground state wave function and the $\ell = 2$ wave function through the dipole operator.

$$O(t) = i\langle\Psi_{\ell=2}(\mathbf{r}, t)|\hat{d}|\Psi_{\ell=0}(\mathbf{r}, t)\rangle \quad (12)$$

We optimized to a two photon transition that is why the overlap function has double periodicity. The vertical line between the field strength and the overlap shows $\pi/2$ phase shift.

When the electric field strength has a local minimum or maximum then the overlap function's carrier oscillation has a zero transition crossing the envelope function. Due to the different time and amplitude scale of the envelope and carrier oscillation this effect is hard to see.

Finally, figure 3 presents the electric field strength of the neutralizing pulse together with the corresponding electron current function. Contrary to the optimized pulse the current has local maximum when the envelope reaches its minimum and vice versa. The system decays with growing field strength. This property of the current can be explained as an off-resonant dynamics, field strength and electron movement. The beat oscillation comes into play again $\tau_{beat} \approx 50/3 \approx 16.6$ a.u. which is hard to identify because $\tau_{\omega_1-\omega_3} = 19.03$ a.u. and $\tau_{\omega_2-\omega_3} = 13.96$ a.u. No unambiguous correlation could be found between the second quick oscillation of the current and the laser field. It is important to mention that different optimized and neutralizing pulses and the corresponding currents were examined from the last generation and all showed the same properties as analyzed above.

IV. SUMMARY AND OUTLOOK

We presented coherent control calculations for the spherically symmetric box potential in short intensive XUV laser pulses. With the help of a GA we maximized and minimized two-photon non-resonant probabilities. We found that the center-of-mass of the electron current is highly correlated with the envelope of the exciting laser pulse. The field of optimized laser pulses force the quiver motion on the electron wavepacket in-phase with the envelope of the pulse. However, and neutralized pulses force the electron to follow a motion which is out-of-phase relative to the envelope.

It is basically possible to implement our method for many-electron atoms too, to investigate and control ionization processes. Such works are in progress. Experimental coherent control experiments are carried out with visible or infrared lights nowadays. Our calculations were done in the hope that the rapid development of laser technology will make such schemes realizable in the near future. Some results of first pioneering experiments are already available [4] with tabletop laser systems, moreover such control schemes are also applicable in free-electron-laser beam-lines delivering femtosecond XUV pulses.

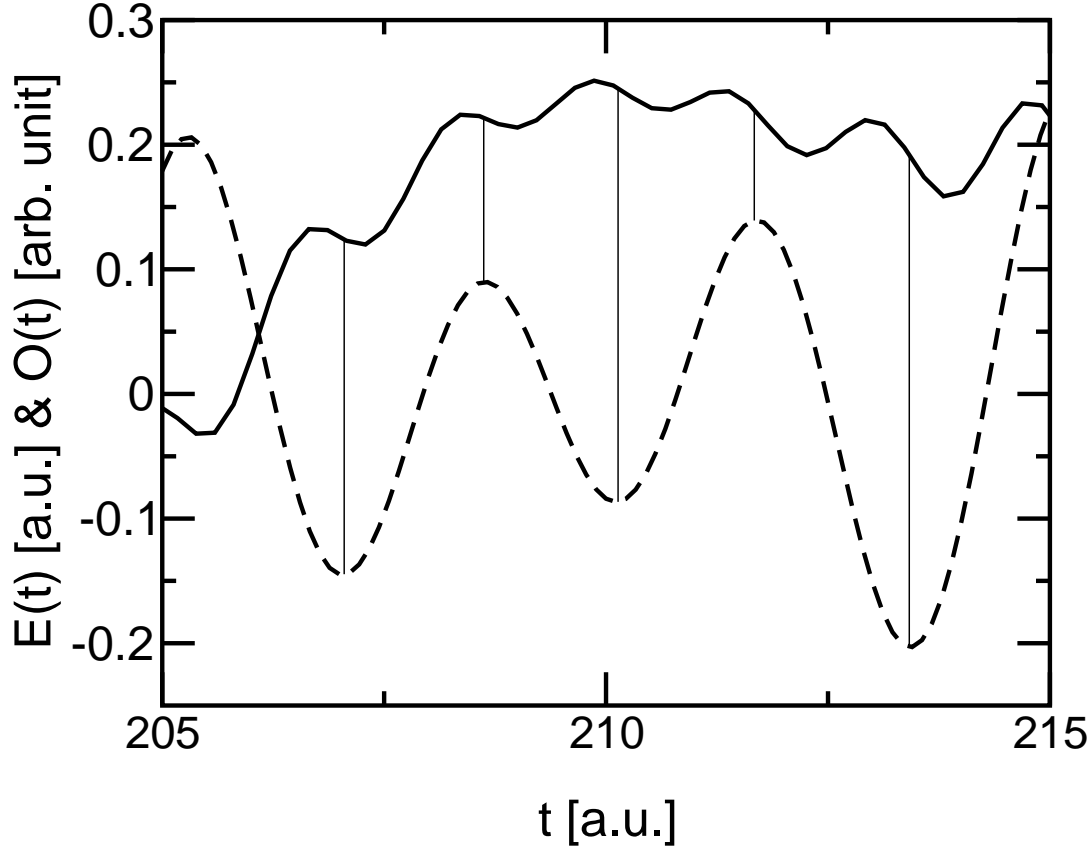


FIG. 2: The $\pi/2$ phase shift between the optimized time-dependent wave function overlap $O(t)$ (solid line) and the electric field strength $E(t)$ of the laser pulse (dashed line)

Acknowledgments

We thank Prof. A. Becker and Prof. J.M. Rost for fruitful discussions and constructive ideas. This work was supported by OTKA (Hungarian Scientific Research Found) F60256 and T048324.

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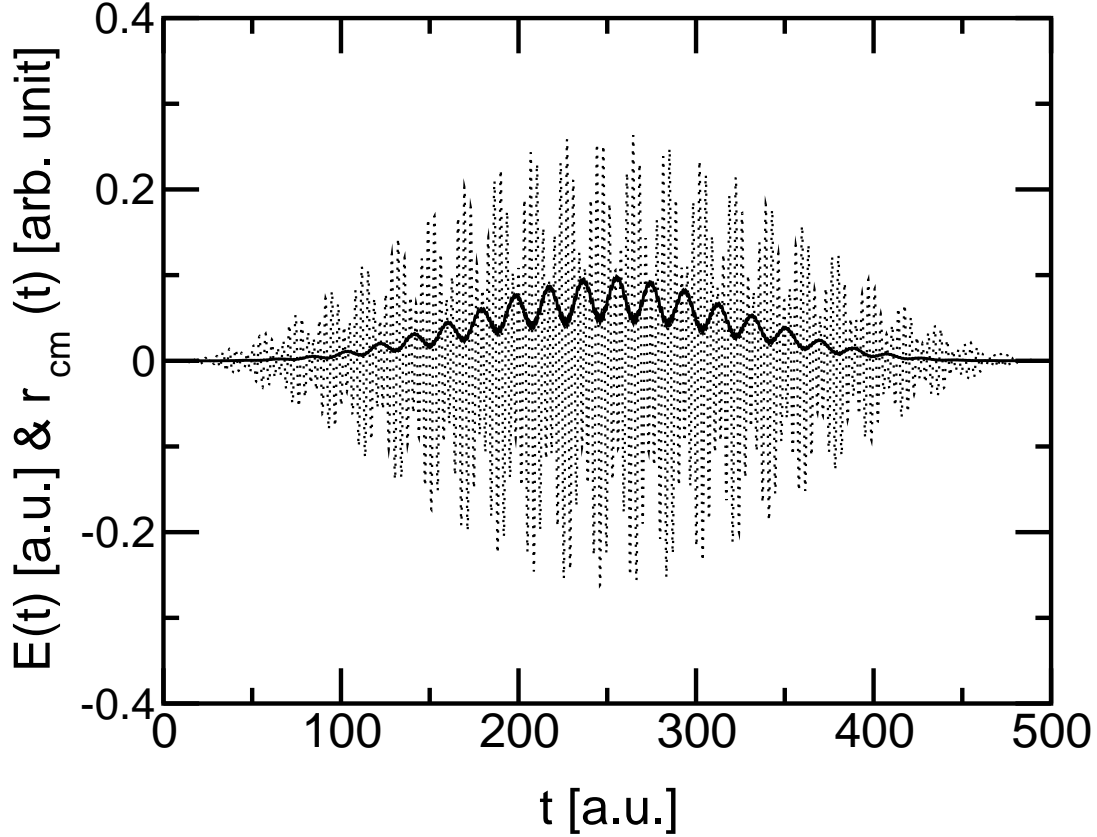


FIG. 3: The electric field strength $E(t)$ of the neutralized pulse (dashed line) with the corresponding center of mass of the electron current $r_{cm}(t)$ solid line)

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